

Title: Carbon Ionic Conductors for Use in Novel Carbon-Ion Fuel Cells

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1. Abstract

Program Introduction: Rational and Objective

Solid ionic electrolytes are a major concern in fuel cell development, but only a few compounds are known to be superionic. The fluorite structure, in particular, has been the basis for several superionic conductors of F^- , I^- , and O^{2-} ions. A number of ionic carbides exist at high temperature in the cubic fluoride structure. The stabilization of these cubic ionic carbides at room temperature or the discovery of high ionic conductivity in other ionic carbides would open the possibility of developing novel fuel cells based on carbon-ion mobility rather than the oxygen-ion mobility that is the basis for current solid-oxide fuel cells.

Accomplishments Achieved during the Current Period of Performance:

Rare earth carbides of the form ReC_2 (where Re refers to any element of the lanthanide series) are being investigated as potential super-ionic membrane materials. These compounds have the fluorite structure when they are above their transition temperatures, which vary from 350 °C (EuC_2) to 1450 °C (LuC_2). The carbon atoms in these compounds reside as anions in tetragonal positions equivalent to the positions of the mobile ions, F^- and O^{2-} in the known superionic conductors CaF_2 and $Zr_{0.8}Y_{0.2}O_2$. In addition, aluminum carbide, Al_4C_3 , has also been produced and is being investigated as another potential carbon ionic conductor.

These ionic dicarbides have been synthesized by reacting mixtures of the appropriate rare earth oxide and amorphous ^{13}C under vacuum at high temperatures ($>1600\text{ }^{\circ}\text{C}$), using a number of newly developed synthesis techniques. The aluminum carbide samples have been produced by reacting ^{13}C directly with aluminum at high temperatures ($>1500^{\circ}\text{C}$) with an argon overpressure of ~ 1 atmosphere in an induction vacuum furnace, and then hot pressing at a temperature of 1100°C and a pressure of 350,000 psi to form dense polycrystalline pellets. Powder x-ray diffraction has been used to confirm the crystal structure of all synthesized compounds.

Plans for the Remaining Period of Performance:

During the remainder of the contract period coatings of ^{12}C will be prepared on the surfaces of the cubic stabilized ionic carbides as well as on the aluminum carbide by vacuum arc-evaporation from normal carbon. The coated samples will be heated at $850\text{ }^{\circ}\text{C}$, $950\text{ }^{\circ}\text{C}$, and $1150\text{ }^{\circ}\text{C}$ in a custom-built high vacuum furnace to allow the ^{12}C to diffuse into the bulk ^{13}C sample. This procedure, preparation of the potential ionic carbide material using ^{13}C rather than normal carbon is necessary because ^{13}C sputtering targets or ^{13}C rods for arc evaporation are unavailable and could not be fabricated. Using a secondary ion mass spectrometer (SIMS), the concentration of ^{12}C and ^{13}C profiles in the bulk material will be measured by determining the distribution profile of ^{13}C with depth as a function of time and temperature of heating. These measurements will then be used to determine a diffusion coefficient for carbon in these ionic carbides as a function of temperature.

2. List of Published Journal Articles, Completed Presentations and Students Receiving Support from the Grant

Papers published since contract inception:

None

Students Supported Under this Grant:

Paul A. Klenk, graduate student in the Department of Mechanical Engineering and Materials Science, Duke University.